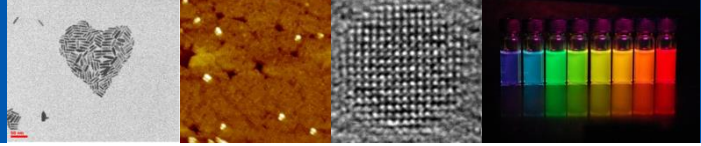

Colloidal quantum dots as single photon emitters

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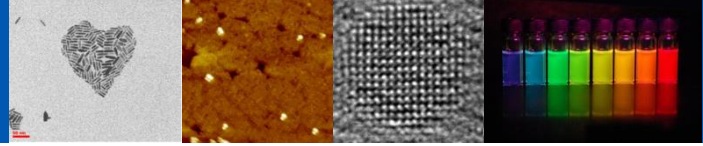
1. Introduction



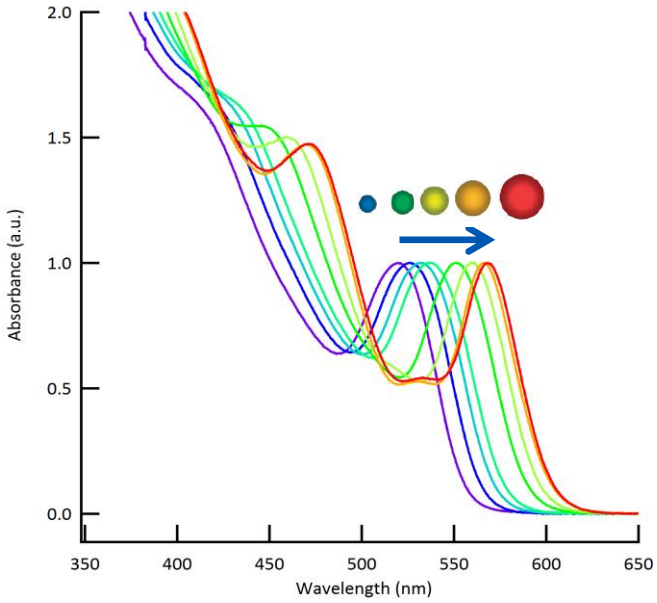
Semiconductor nanocrystals

→ High tunability

→ Different materials available (CdSe, PbSe, CdS, ZnS,…)

→ Influence of size (quantum confinement)

→ Possibility to synthesize core/shell structures



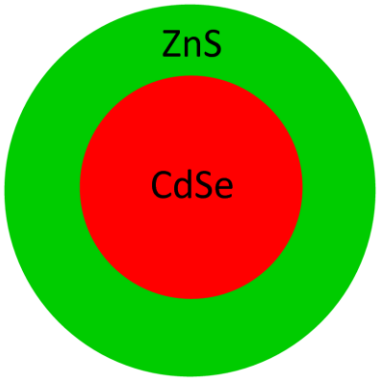
Can we use them as single photon sources ?

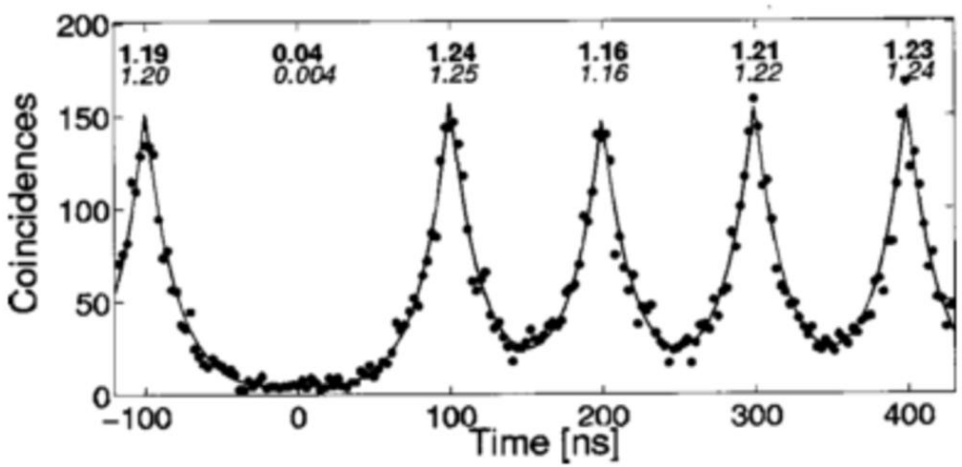
1. Introduction

Single photon emission for quantum optics

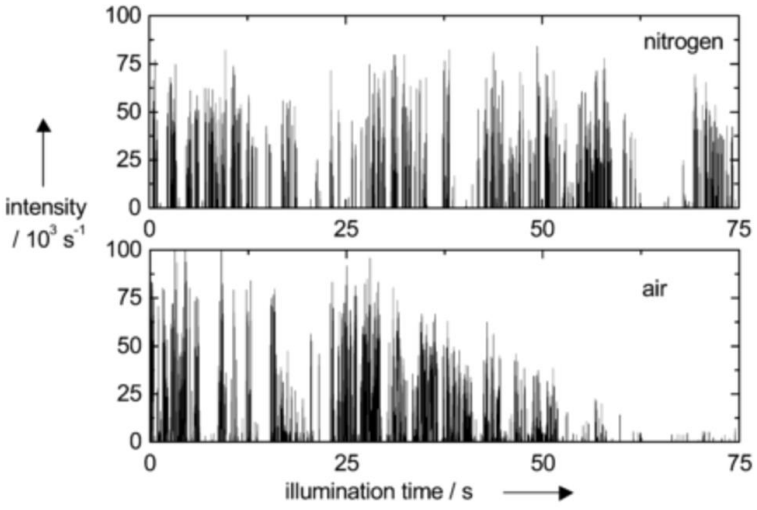
→ Single photon emission demonstrated in CdSe / ZnS QDs

*Brokmann et al., APL, **85**, 5, 2004*





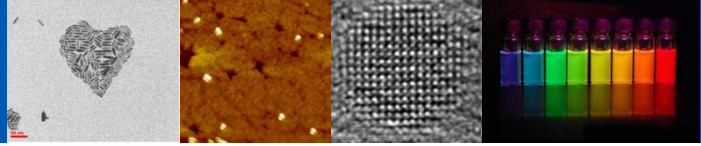
→ Blinking behaviour, i.e. succession of ON and OFF states



Objective : Deterministic photon source

No photon on demand behaviour

*Van Sart et al., Chem Phys Chem, **3**, 10, 871-879, 2002*



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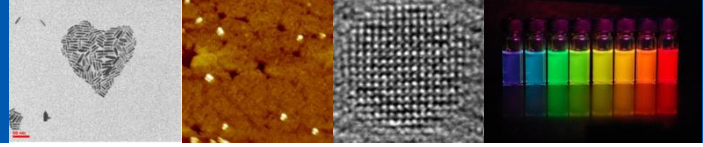
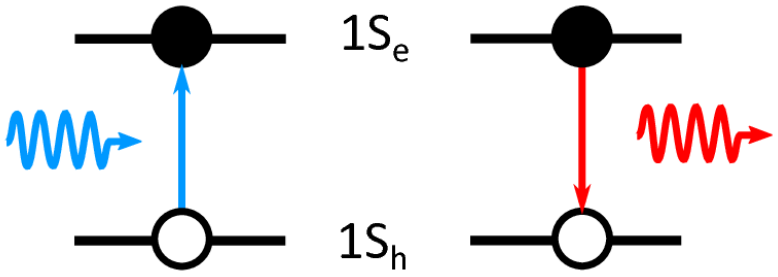
2. Single QD as single photon emitter

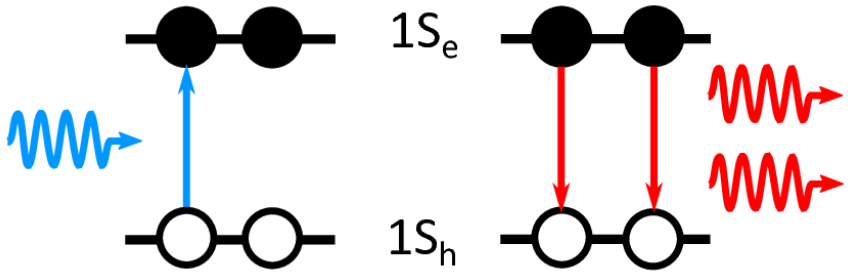


Impact of Auger recombination process

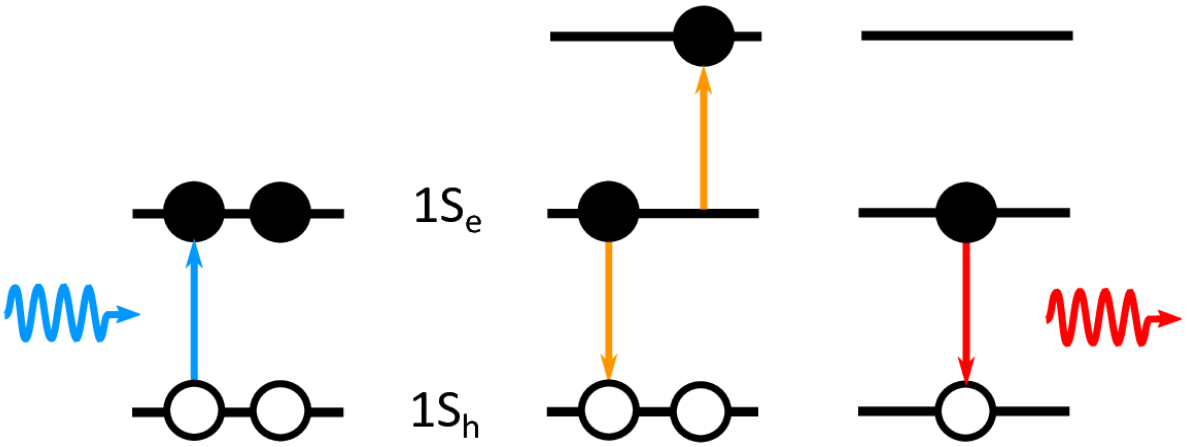
→ What is responsible for good single photon emission properties in QDs ?



Single exciton emission



Bi-exciton recombination : emission

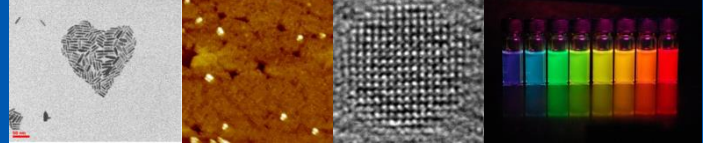


Bi-exciton recombination : Auger, non radiative

Strong Auger effect:

Efficient single photon emitter

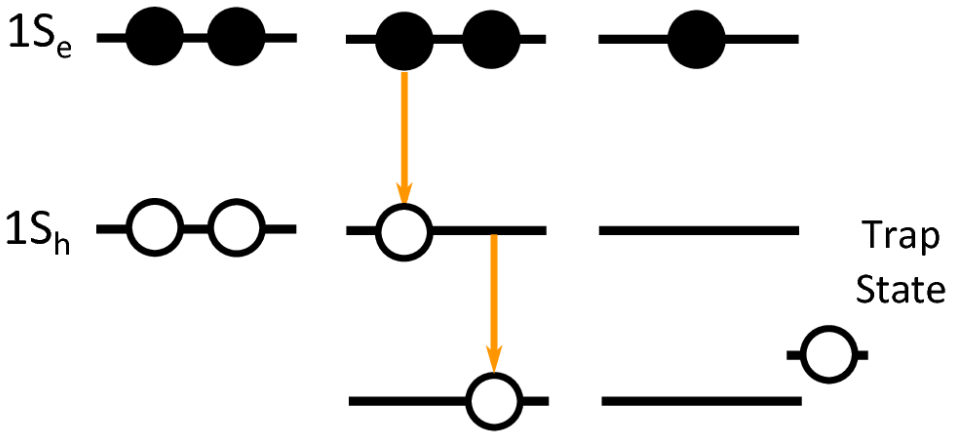
2. Single QD as single photon emitter



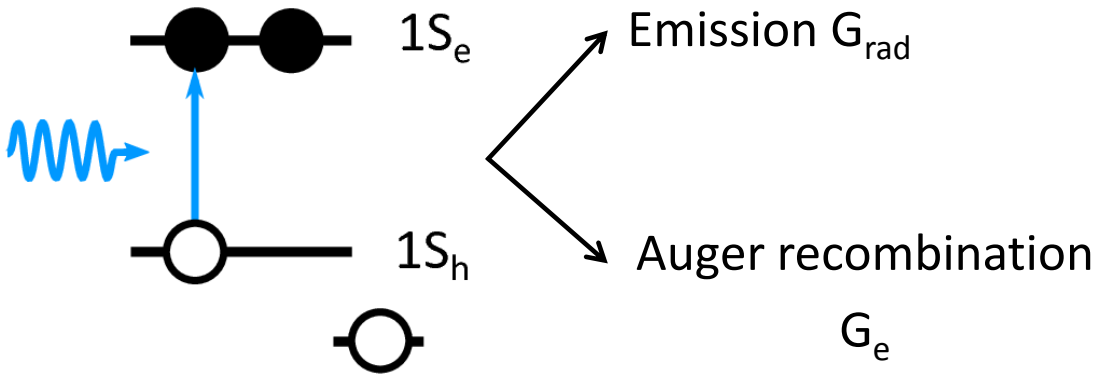
Link between Auger process and Blinking behaviour

→ Efficient Auger recombination rate : Generation of hot charge carriers

→ Trapped charge : Charged QD



→ Recombination of charged state (trion) : Radiative and non-radiative pathways



$$G_{X+e} = G_e + G_{rad}$$

In CdSe/ZnS : $G_e >> G_{rad}$

No emission in charge QD

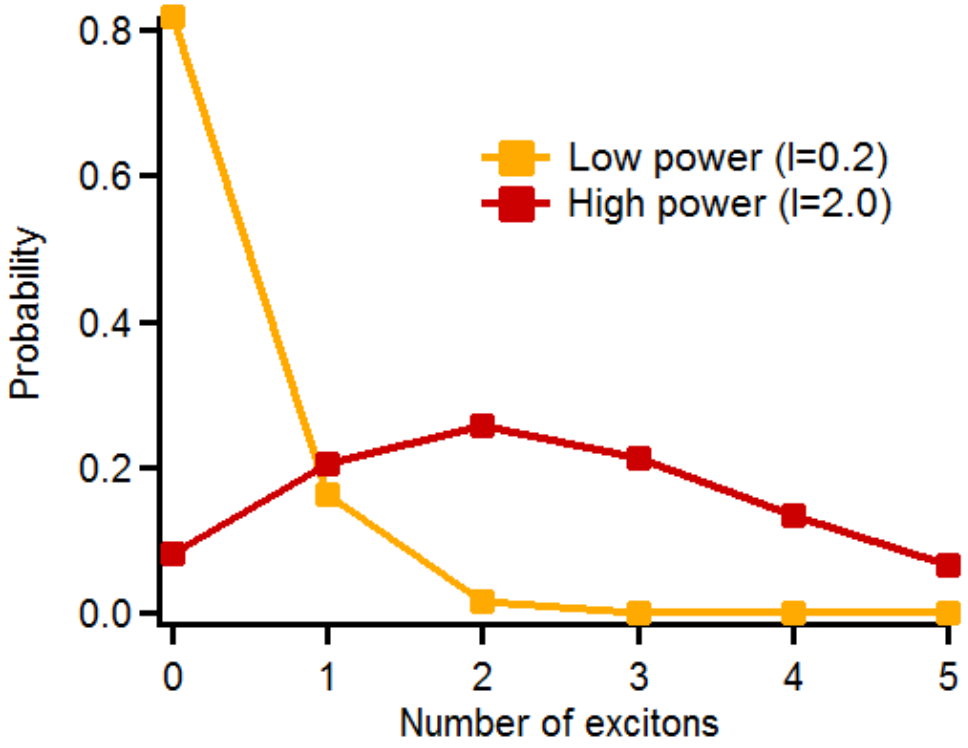
Blinking

2. Single QD as single photon emitter

Necessary control of Auger recombination rate

- → Pulsed excitation
- → Number of exciton is Poissonian

- → Low fluence :
  - → No bi-exciton
  - → No photon on demand
- → High fluence :
  - → Almost photon on demand
  - → Creation of multi-excitons



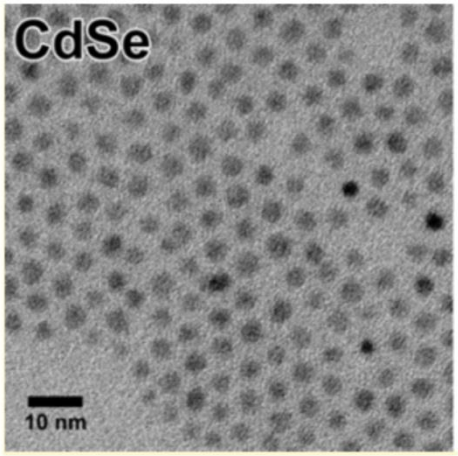
High excitation power needed

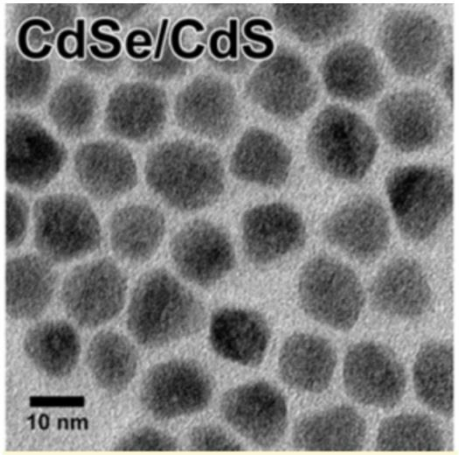
Auger process gives single photon emission and blinking behaviour

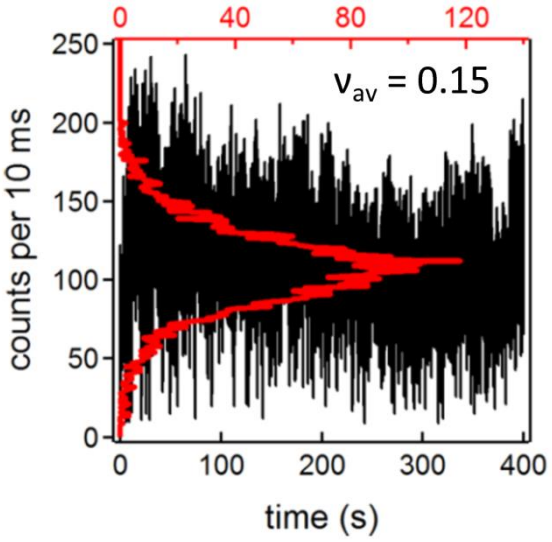
2. Single QD as single photon emitter

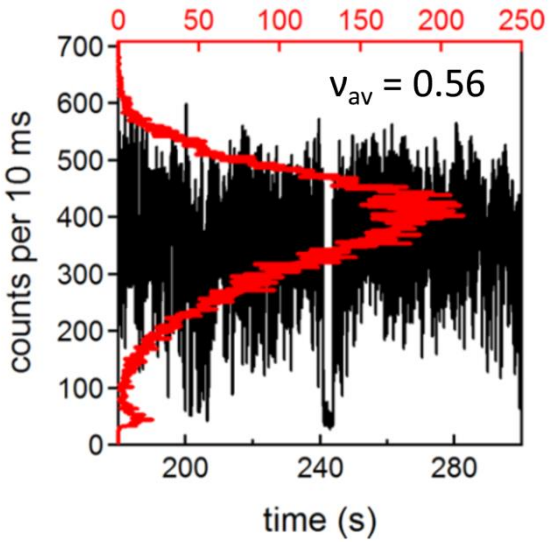
Non-blinking CdSe / CdS

*Cirillo et al., Chem. Mater., **26**, 2, 2014*





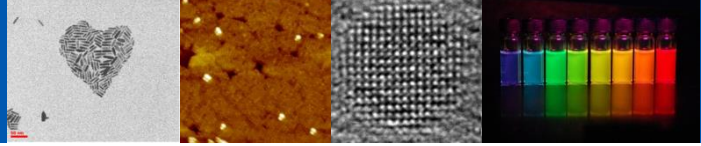




→ High quantum yield

→ Non blinking

Usable for single photon emission ?



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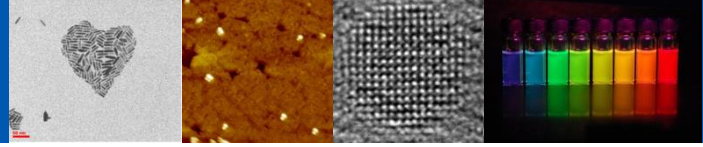
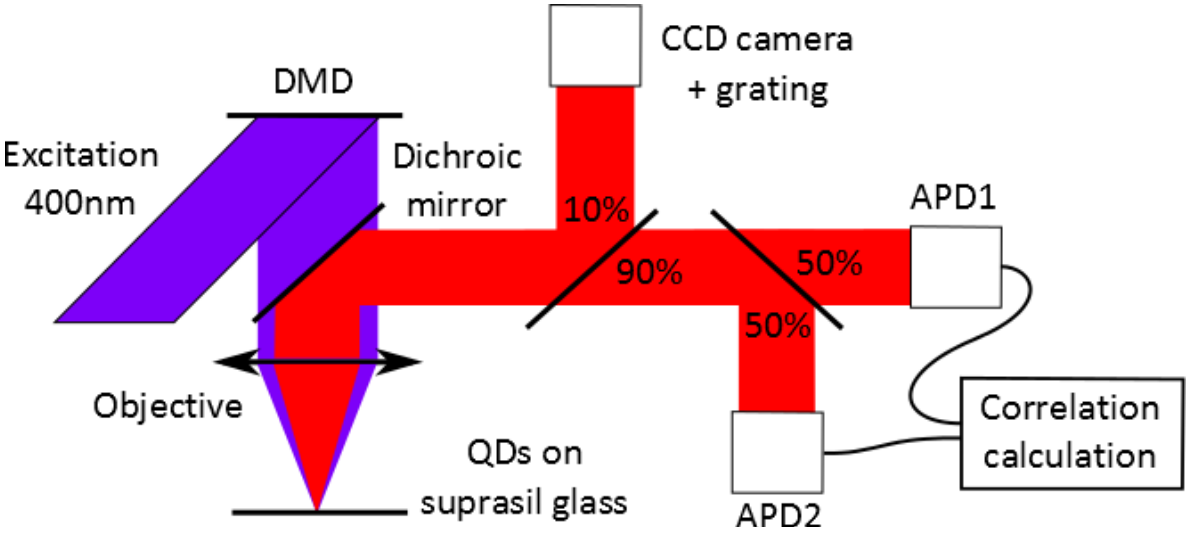
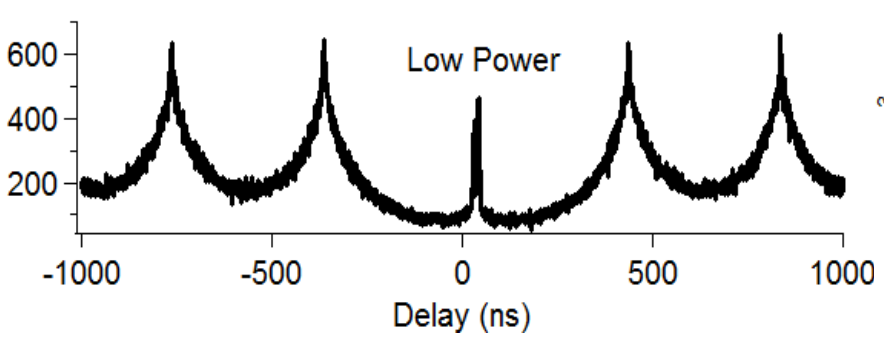
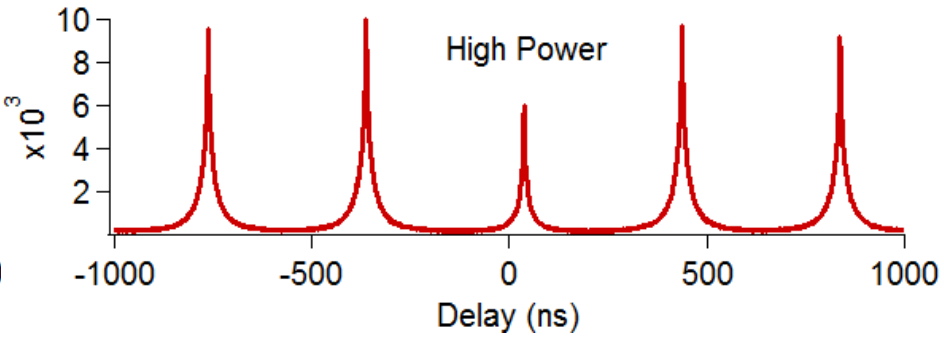


Experimental setup and emission correlation measurement

→ Measurement on single QD with fluorescence microscope and HBT detection







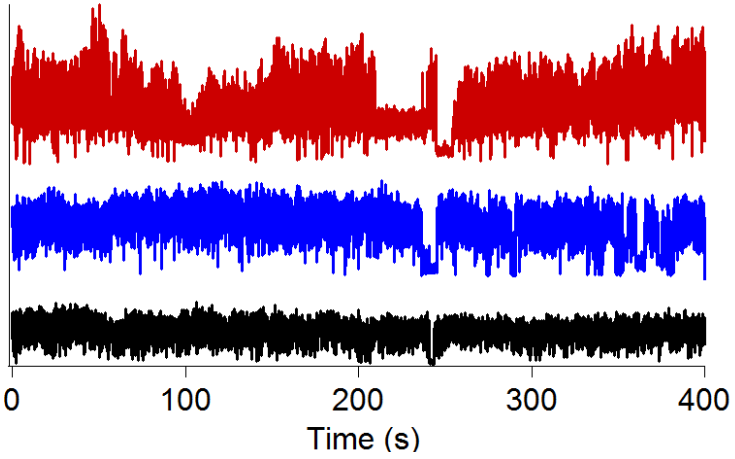
→ Good single photon emission in giant CdSe / CdS QDs at low excitation power

→ Better understanding of their properties is required

3. Results and discussion

Evolution of blinking trace with excitation power

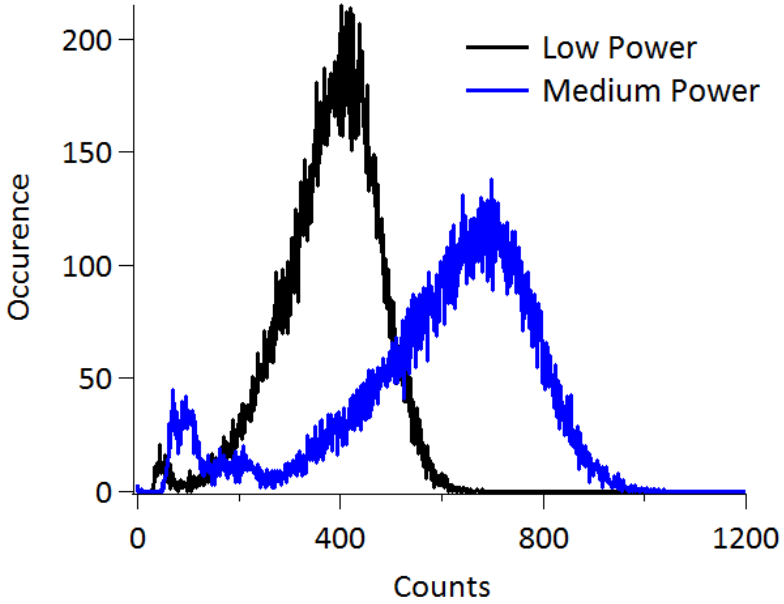
→ One giant CdSe / CdS QD under different excitation power

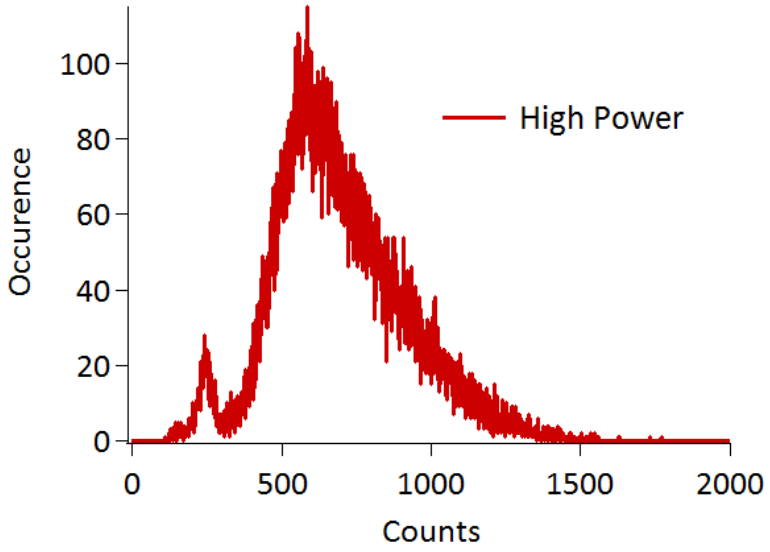


→ Non blinking even at high power

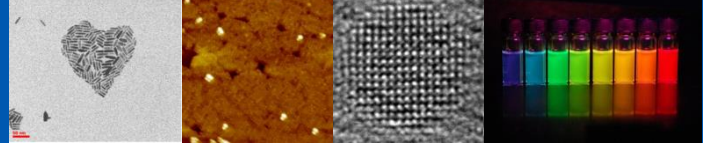
→ Distribution changes at high power

QD in different states with different quantum yield ?

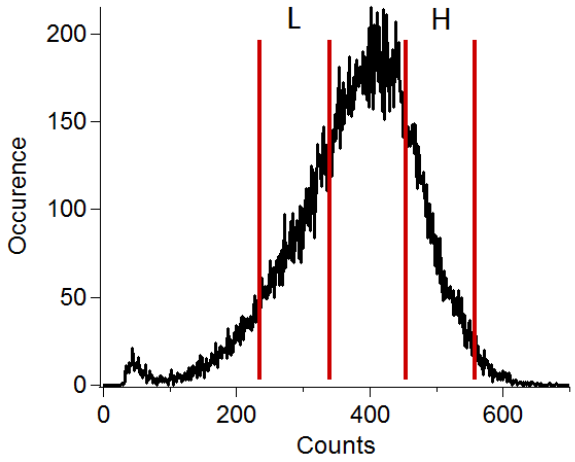


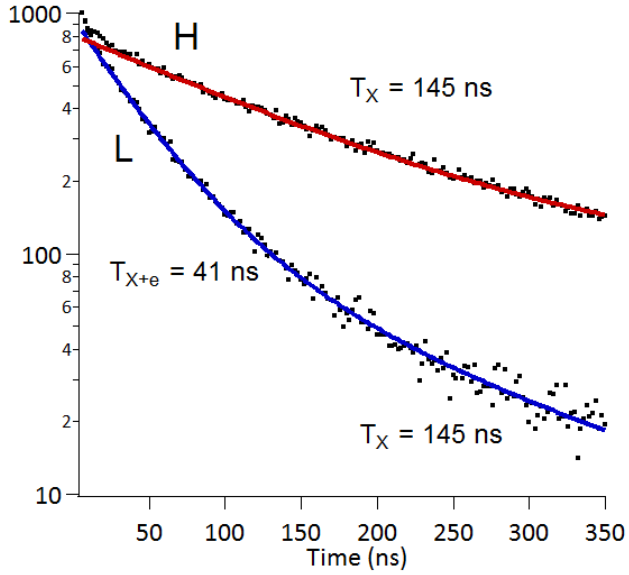


3. Results and discussion

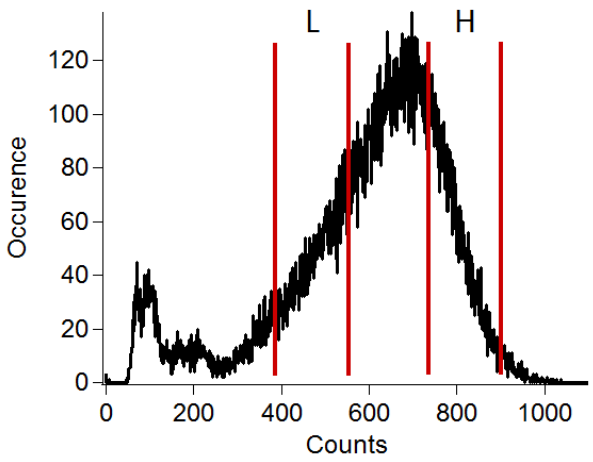


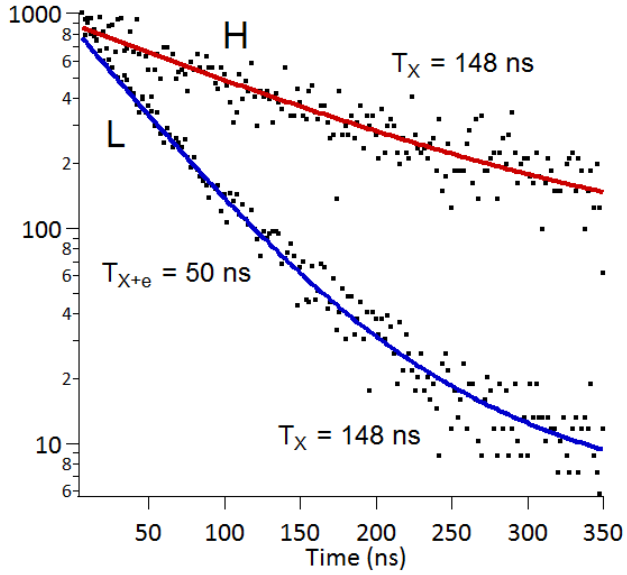
Low power



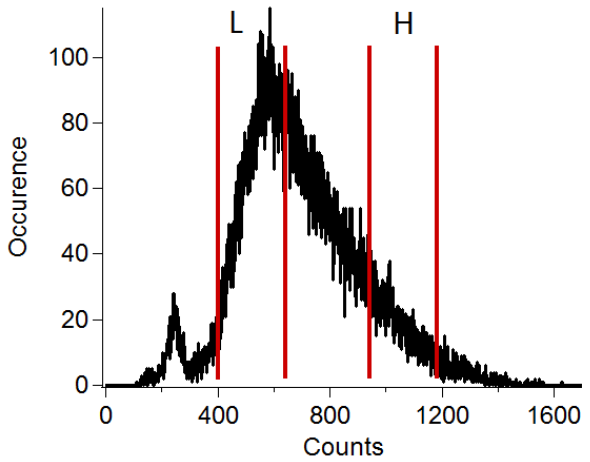


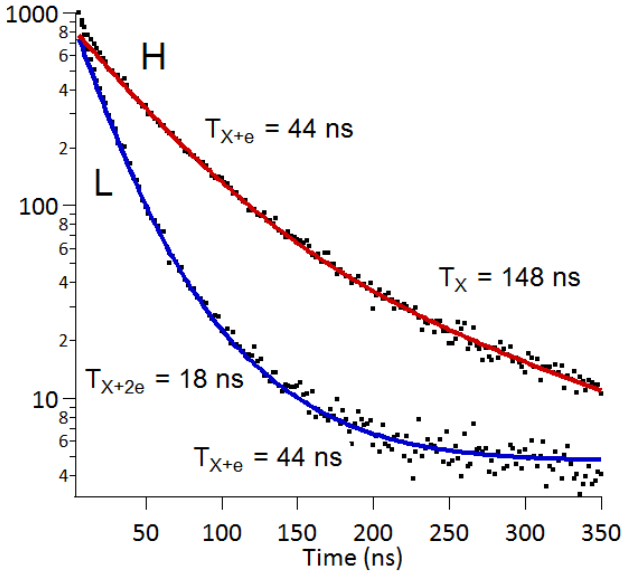
Medium power





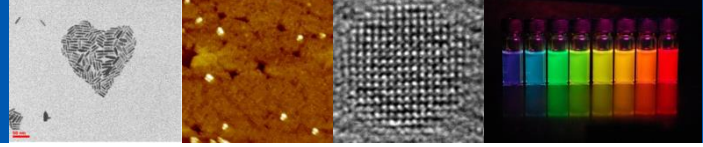
High power





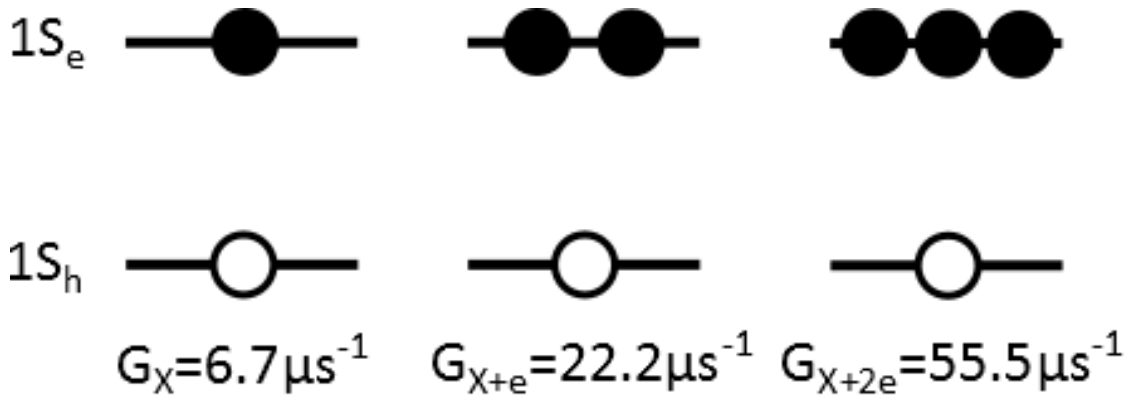
→ QD can be in different states : Confirmed by the different lifetimes

3. Results and discussion



Lifetime in different charged state ?

→ QD can take different charged state



Trion

$$G_{X+e}=G_{rad}+G_e$$

$$G_{rad}=2G_X$$

$$G_e=G_{X+e}-G_{rad}$$

→ Recombination of biexciton

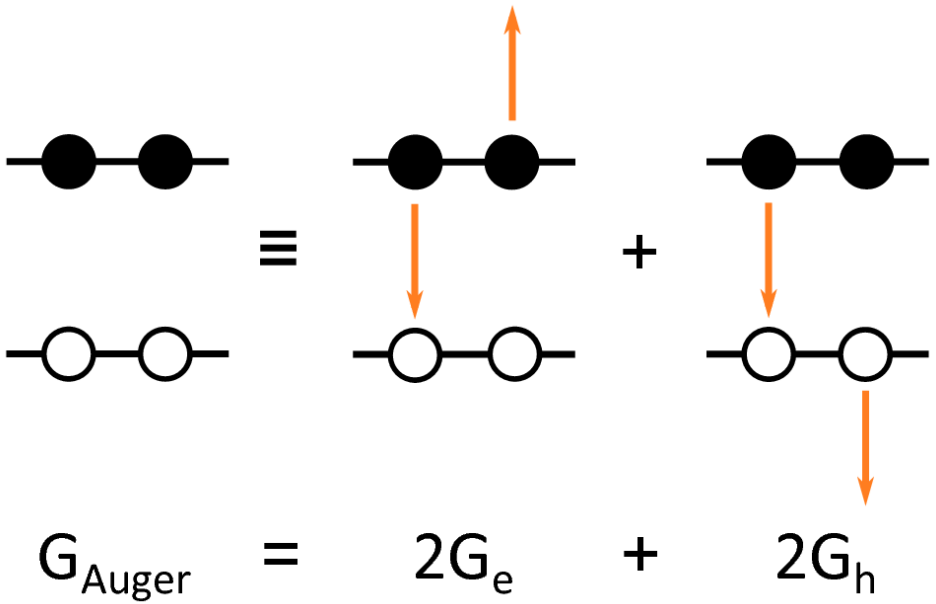
→ Two pathways

→ Electron channel : G_e

→ Hole channel : G_h

G_e, G_h, G_{Xrad} known

Calculate correlation function in various charged states



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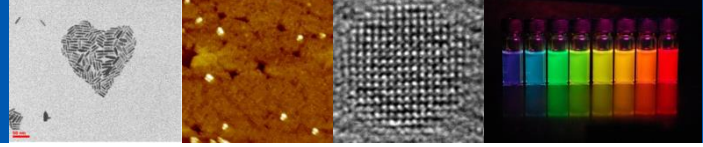
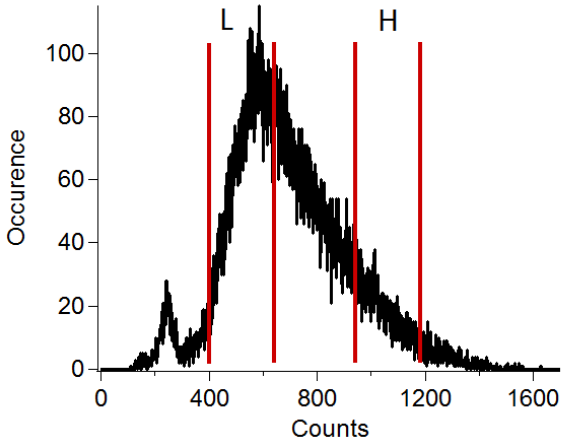


Calculation of correlation function

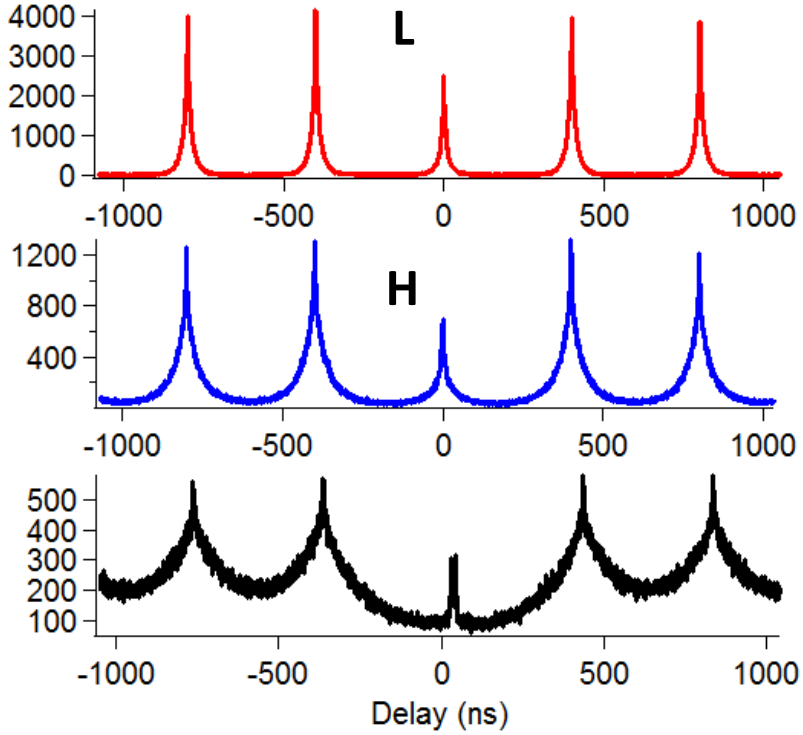
→ QD in a succession of charged state

→ Each state shows a different correlation function

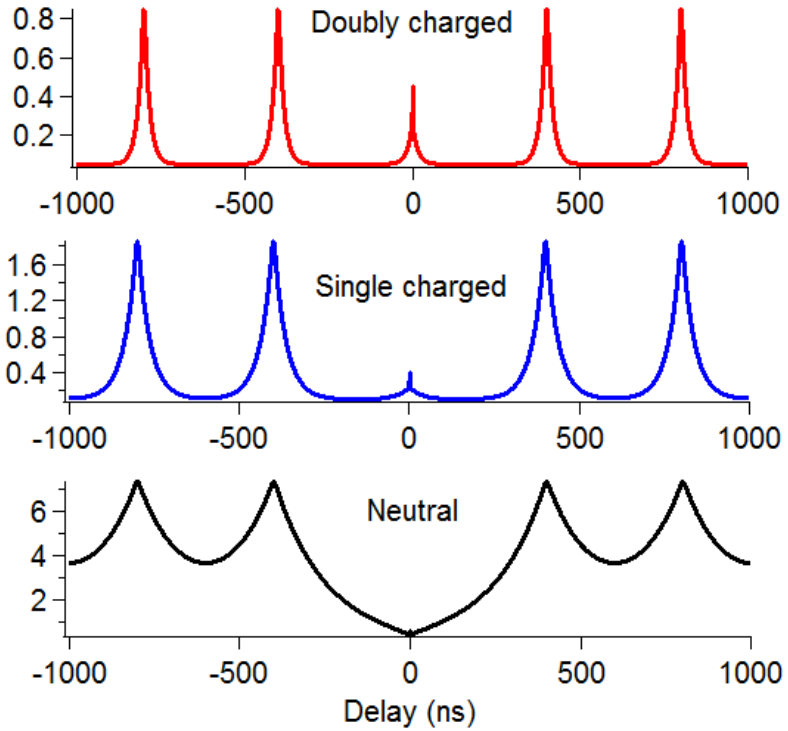
→ Evolution reproduced with calculation



Experimental



Calculated



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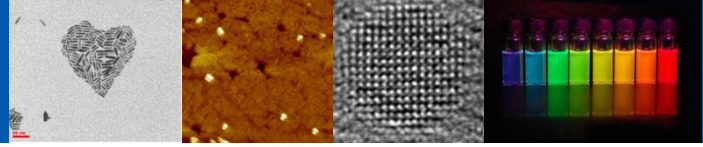
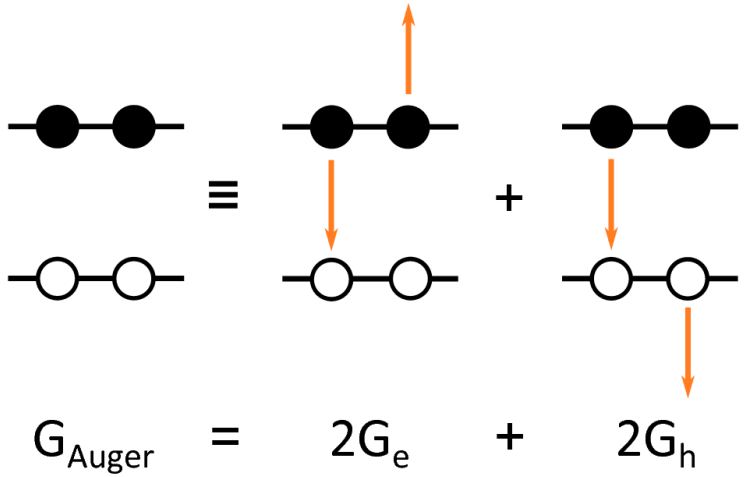


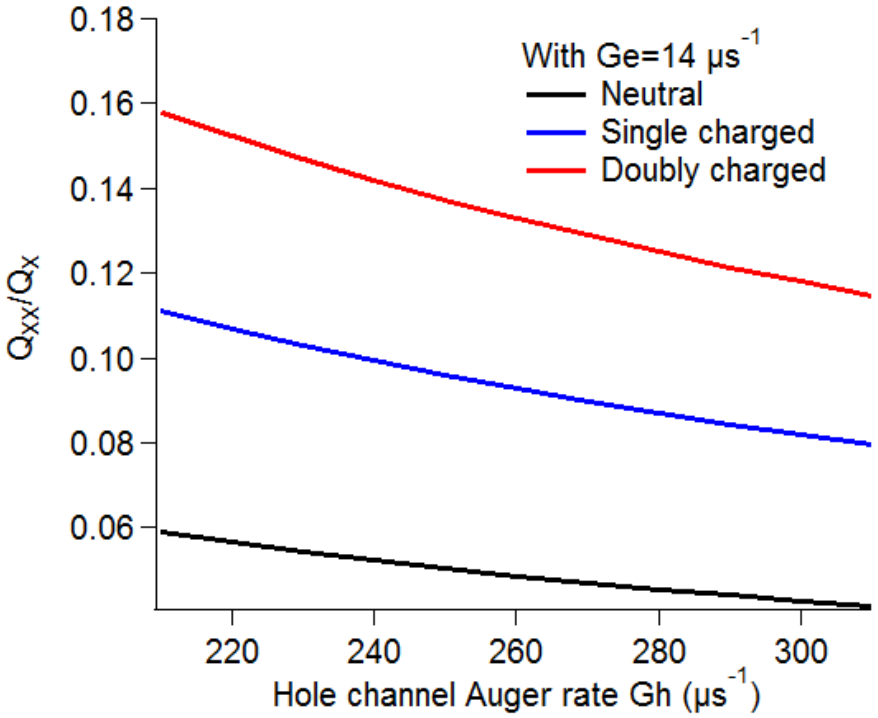
Influence of different Auger rates

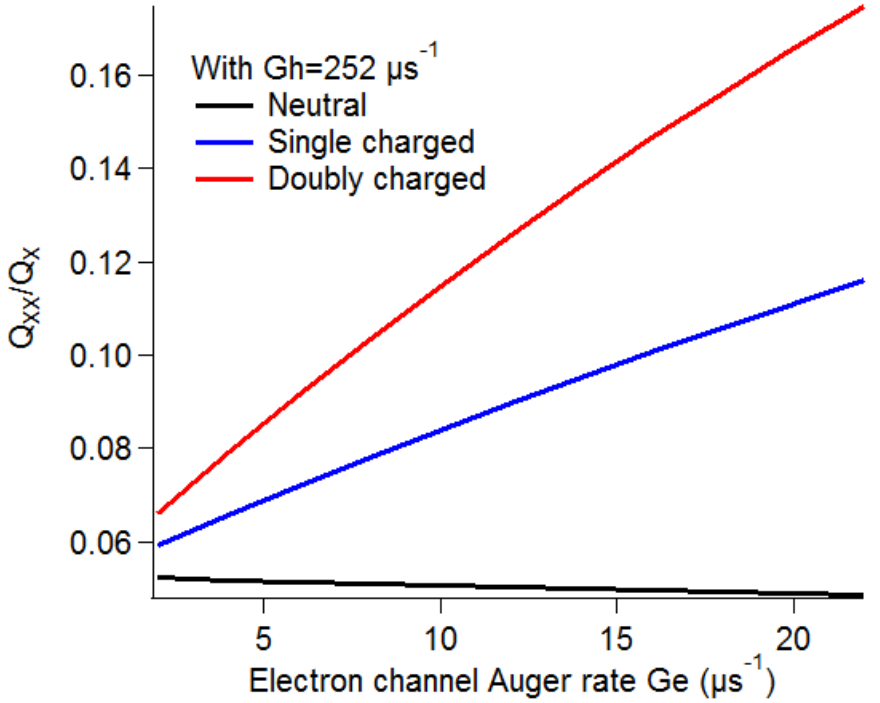
→ Quantum yield of biexciton less affected by additional charge in QD

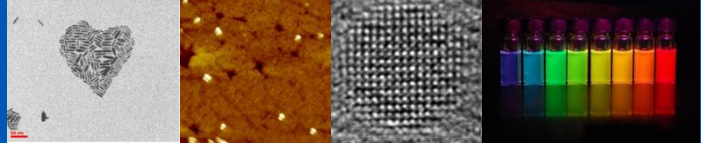
→ Responsible for degradation of single photon emission performances

→ Very low G_e and very high G_h recommended









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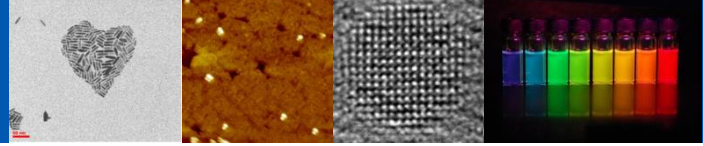


Colloidal QD as single photon emitter

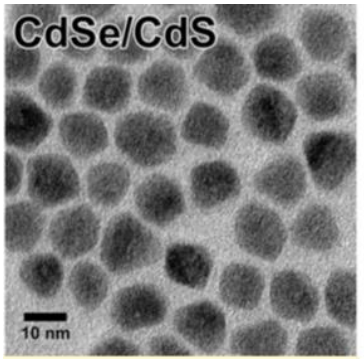
→ Achieve deterministic photon source with single colloidal QD

→ Must be non-blinking

→ Single photon emission must be preserved at high excitation fluence

Non blinking giant CdSe\CdS QD

→ High quantum yield, non-blinking CdSe\CdS synthesized with flash method

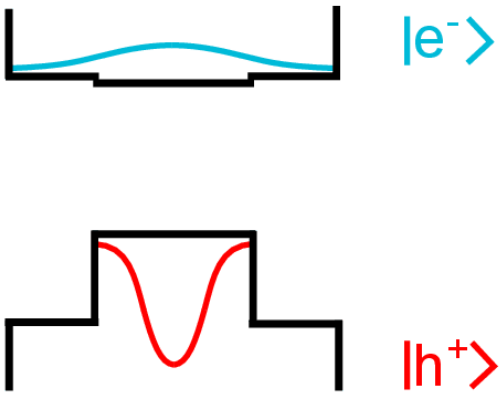


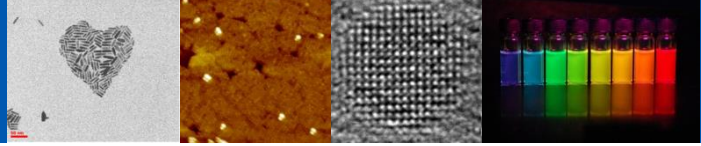
Control of Auger recombination

→ Reduce G_e : increase size of shell, alloying at interface

→ Increase G_h : decrease size of the core, abrupt interface

→ Control of surface properties and crystallinity





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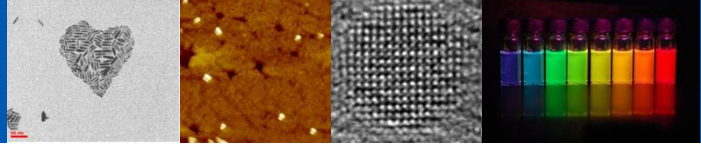

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